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MERIDIONAL DISTRIBUTIONS OF DEUTERIUM IN ATMOSPHERIC WATER VAPOUR BETWEEN TROPICAL AND SOUTHERN POLAR LATITUDES

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Abstract: The temporal and spatial variations in the deuterium/hydrogen isotopic ratio of tropospheric water vapour in air masses circulating above the Southern Ocean and the Antarctic continental ice sheet were investigated during the summer season of the 36th Japanese Antarctic Research Expedition (JARE-36) from November 1994 to March 1995. The results of the deuterium analyses of air moisture samples are presented together with water, air and dew point temperatures as functions of latitude. A strong relationship between isotopic composition and latitude as well as corresponding temperatures was found in the region from 20° N to 60° S. Specific circumpolar, meridional and vertical atmospheric circulations and katabatic wind fields dominate in the Antarctic region, hence temperature dependences of the deuterium distribution in Antarctic water vapour are not significant during the short austral summer observation period. The deuterium distributions there are also different from the altitude and latitude effects which are typically for lower latitudes. Furthermore, it is shown that the isotopic composition of air moisture collected at higher latitudes is strongly affected by condensation and precipitation processes during transport, rather than by the evaporation temperature in the source region.

1. Introduction

Stable isotopes of water are not only useful indicators of climatic changes in the past, they also provide insight into today's changing climate. They are particularly useful in studying the water cycle and the interactions between ocean, atmosphere, snow and ice surfaces. The stable isotope compositions of atmospheric water vapour and precipitation are influenced by several climatic and geographic factors, often termed altitude, amount, continent, latitude, and season effects (*e.g.* HOEFS, 1987). Widely discussed is also the importance of the origin of atmospheric water vapour (KOSTER *et al.*, 1992). All these effects are more or less related to temperature, as temperature controls isotopic fractionation during phase transitions. Furthermore, changes in the isotopic composition of atmospheric water vapour and in precipitation are caused by transport, mixing, exchange, condensation and precipitation processes. Thus, the final isotopic composition in falling and accumulated snow depends on temperature and

several dynamic processes. Much time and effort has been invested in reconstructing paleoclimate by isotopic ice core analysis using an empirical relationship to mean surface temperature values (*e.g.* MAYEWSKI *et al.*, 1996). However, less effort is being put into understanding the reasons behind the variation in the primary isotopic inflow to polar ice caps by atmospheric water vapour. Often only surface snow, snow pits or firn cores are used to assess the isotopic composition of recent precipitation. Sampling of falling snow or even air moisture is very rare in polar regions.

Participation in the summer party of the 36th Japanese Antarctic Research Expedition (JARE-36) from November 1994 to March 1995 presented an opportunity to begin an investigation into temporal and spatial variations of the deuterium/hydrogen isotopic ratio of tropospheric water vapour in air masses circulating above the Southern Ocean, and at several elevations on the Antarctic continental ice sheet. This study focuses on the latitudinal variation of the isotopic composition of atmospheric water vapour between tropical latitudes and the Antarctic continent during the southern summer season.

2. Sample Collection and Analysis

The collection of atmospheric water vapour was carried out using newly developed air moisture sampling equipment (SCHWARZ, 1996). The instrument contains a changeable magazine with 12 sampling traps filled with granulated molecular sieve for moisture adsorption. It is controlled by a microprocessor, and takes samples with the aid of 14 magnetic valves and a small vacuum pump according to a given time schedule. In the laboratory, the collected water is desorbed quantitatively under vacuum and heating, so that no isotope fractionation is possible during this procedure. This is followed by reduction by hot chromium, which produces hydrogen gas samples ready for isotopic analysis by mass spectrometry.

The accuracy of the mass spectrometer measurements was about 0.5‰ and the reproductivity by repetitions and doubles was lower than 2‰.

The new sampler was operated on board the icebreaker SHIRASE between Tokyo and Syowa Station in November and December 1994 (9 samples), and also between Syowa Station and Sydney in February and March 1995 (7 samples). In Fig. 1 the individual sampling locations and dates are shown for both voyages. At each location, one sample of air moisture was taken, and the air temperature and dew point temperature at 10 m altitude as well as the ocean surface temperature were measured. In addition, during January 1995 several samples for stable isotope analysis were collected at Japanese Antarctic stations (SCHWARZ *et al.*, 1996). Twenty-one air moisture samples were collected at 21 m a.s.l. at the coastal Syowa Station (69.00°S, 39.58°E), and in parallel, using the same type of sampling instrument, at two continental stations, *i.e.*: 12 samples at 2230 m a.s.l. at Mizuho Station (70.70°S, 44.33°E) and 7 samples at 3810 m a.s.l. at Dome Fuji Station (77.32°S, 39.70°E).

The resulting deuterium/hydrogen isotopic ratio R is commonly expressed in terms of the δ notation as follows:

$$\delta D = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000‰, \quad (1)$$

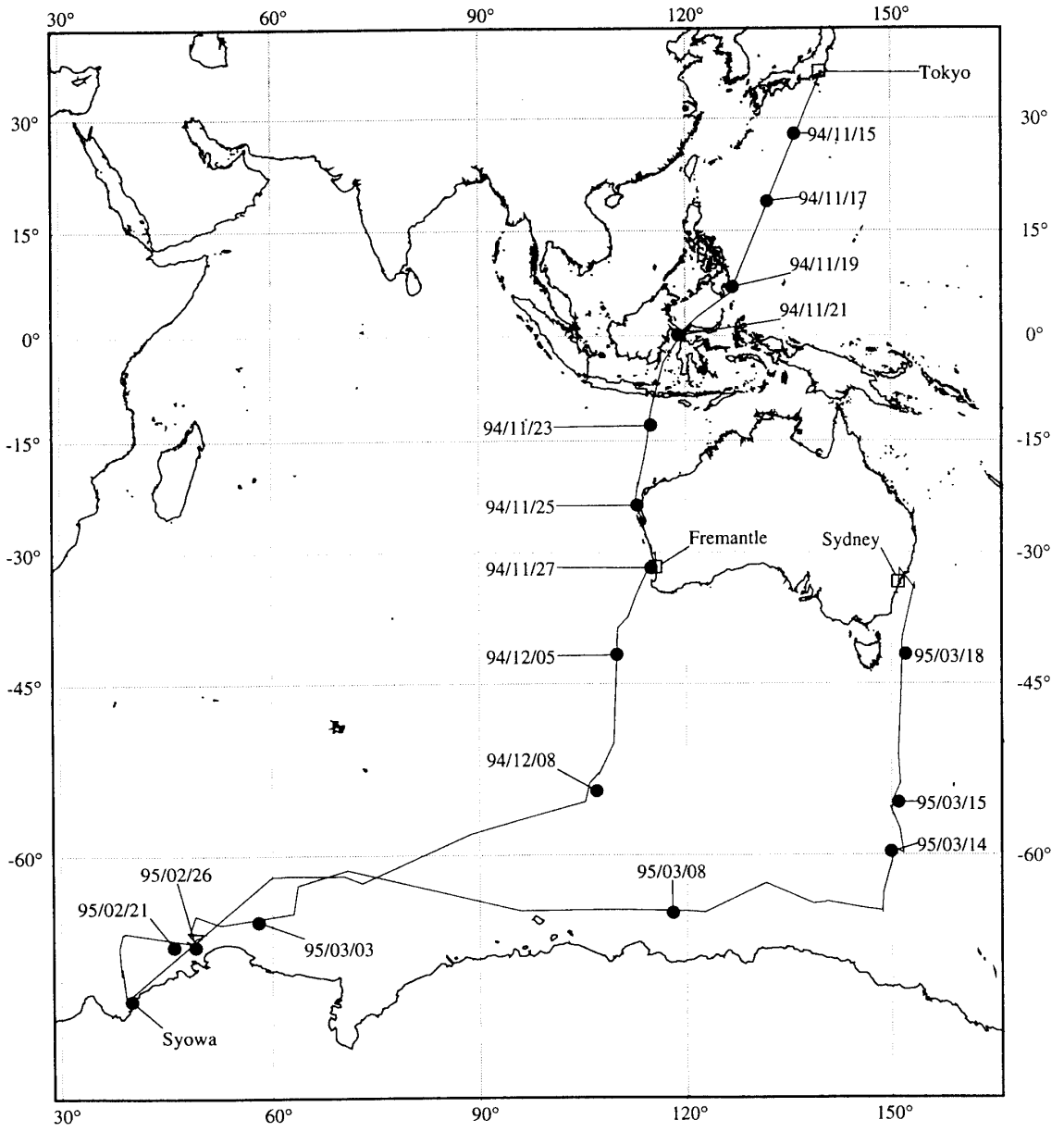

Fig. 1. Route of RV SHIRASE during JARE-36; locations and dates of air moisture sampling.

i.e. in relation to the isotopic standard SMOW (standard mean ocean water). The experimental δD values for the different geographical locations and the average values from Syowa Station for January 1995 are compared in Fig. 2 with the measured temperatures of air, dewpoint and ocean surface water. While the ship-borne data indicate the latitudinal variation for the periods November/December 1994 and February/March 1995, the 69°S data show the means recorded at Syowa Station for January 1995. The mean δD values of all three Antarctic stations together with some theoretical estimations are shown in Fig. 3 for January 1995 and the ship-borne data.

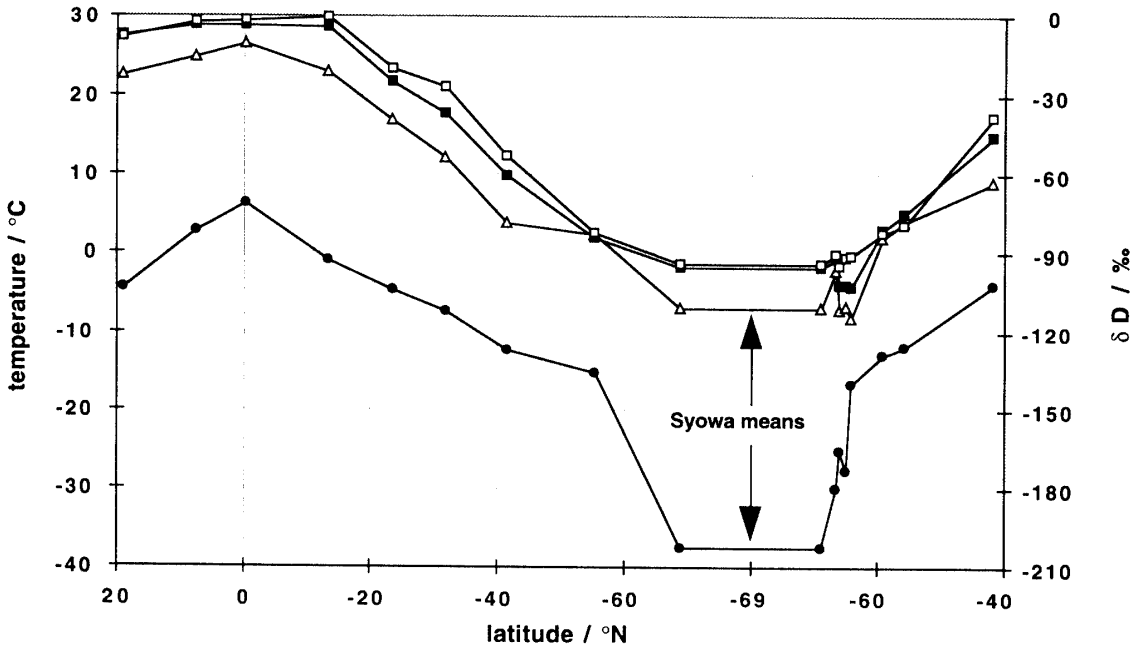

Fig. 2. δD values of air moisture samples (solid circles) as well as air (solid squares), dewpoint (triangles) and surface water (open squares) temperatures versus latitude.

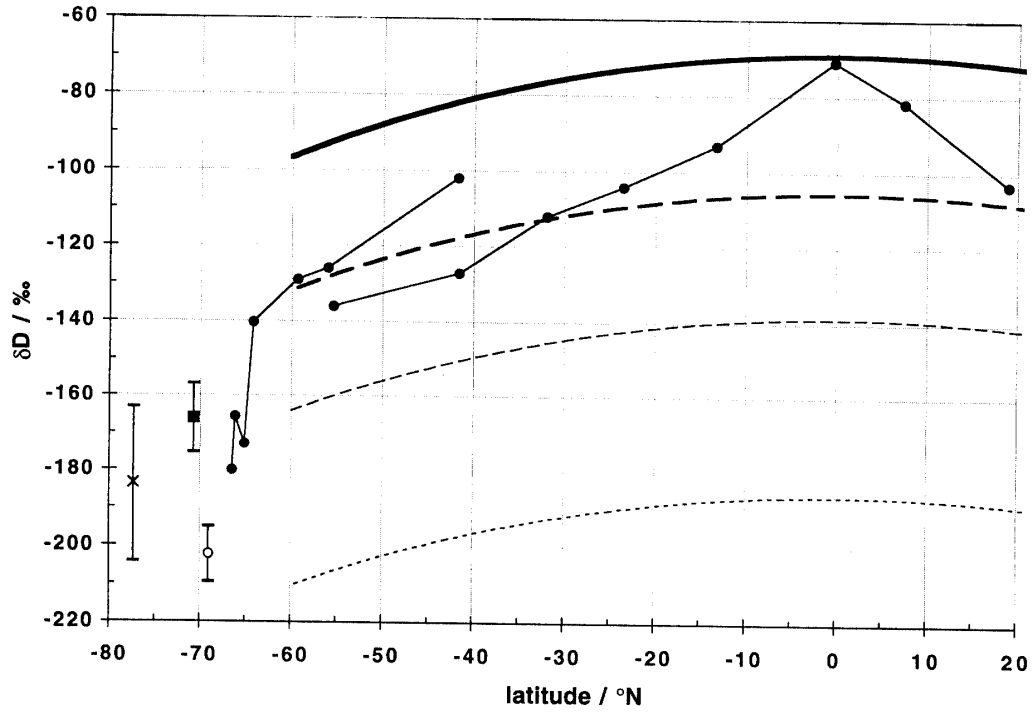

Fig. 3. Measured δD values: ship-borne (solid circles), Syowa Station (open circle), Mizuho Station (solid square) and Dome Fuji Station (cross). δD means are given for the Antarctic stations together with their 95% confidence intervals. Also shown are calculated δD values: $\delta D_{evap.}$ (bold solid line), $\delta D_{vap.}$ for $f=0.7$ (bold dashed line), $\delta D_{vap.}$ for $f=0.5$ (thin dashed line), and $\delta D_{vap.}$ for $f=0.3$ (dotted line).

3. Results and Discussion

The isotopic composition of air moisture shows a strong dependence on geographical co-ordinates and temperatures (Fig. 2). Highest δD values were observed at tropical latitudes. Significantly lower values appear in polar latitudes at lower temperatures. This result is consistent with the latitude and temperature effects discussed by DANSGAARD (1964) and HOEFS (1987). A strong decrease of δD occurs between 60°S and 69°S.

This latitudinal variation in δD is plotted with the January δD means for Syowa Station, Mizuho Station, and Dome Fuji Station, as well as with some estimated water vapour curves in Fig. 3. The lowest δD values were recorded at the coastal region of the Antarctic continent at Syowa Station. At Mizuho Station, located at 2230 m altitude at 70.70°S, the δD values are significantly higher than in the coastal region near sea level (Syowa Station) and correspond to the δD values recorded at sea level near 65°S. The southernmost location at 3810 m altitude at 77.32°S (Dome Fuji Station) again shows lower δD values than those from Mizuho Station. The variability however, is greater at Dome Fuji Station than at Mizuho Station and at Syowa Station.

The observations between 20°N and 60°S are compared with a theoretical estimation of the deuterium/hydrogen ratio in the atmospheric water vapour after evaporation ($\delta D_{\text{evap.}}$) as a function of the corresponding ocean surface water temperatures. According to HORITA and WESOLOWSKI (1994) the temperature dependent liquid-vapour fractionation factor α, defined as

$$\alpha = (D/H)_{\text{liq.}}/(D/H)_{\text{vap.}} \quad \text{or} \quad \alpha = R_{\text{liq.}}/R_{\text{vap.}}, \tag{2}$$

for deuterium, can be approximated by

$$\ln\alpha = 1158.8\,(T^3/10^{12}) - 1620.1\,(T^2/10^9) + 794.84\,(T/10^6) - 161.04 + 2.9992\,(10^6/T^3), \tag{3}$$

with the phase transition temperature T in degrees Kelvin. The $\delta D_{\text{evap.}}$ for evaporation of ocean water with $\delta D = 0‰$, equal to standard mean ocean water (SMOW), is then simply represented by

$$\delta D_{\text{evap.}} = (1/\alpha - 1) \times 1000‰. \tag{4}$$

For the approximation of α a quadratic least square fit function of the measured surface water temperatures was used. The resulting curve is the uppermost one (bold solid line) in Fig. 3. It shows the temperature dependence of the deuterium content in evaporated ocean water. Near the equator, $\delta D_{\text{evap.}}$ coincides with the measured δD value of air moisture. So the δD value of original atmospheric water vapour is explained by evaporation according to eq. (4). It is obvious that the deviation between calculated $\delta D_{\text{evap.}}$ and measured δD values increases with increasing latitude. Quite similar results were obtained during ship cruises by the German research parties of Soviet Antarctic Expeditions (KOWSKI *et al.*, 1982). Hence local evaporation as a

function of temperature does not seem to be the dominant process affecting isotopic ratios in water vapour outside tropical latitudes. These deviations show that condensation and precipitation processes are probably more important for isotopic fractionation within extratropical wet air masses.

Such isotopic fractionation is discussed by DANSGAARD (1964) in terms of Rayleigh type condensation which takes place in an open system. The $\delta D_{vap.}$ values of remaining water vapour can be estimated using the Rayleigh model (HOEFS, 1987) for different water depletion ratios according to eq. (5).

$$\delta D_{vap.} = \{(\delta D_{evap.}/1000+1) \times f^{(\alpha-1)} - 1\} \times 1000‰. \quad (5)$$

For this purpose, a constant condensation temperature of 0°C was assumed for the calculation of α in eq. (3). The value f is the fraction of the remaining water vapour within the air mass after Rayleigh condensation and immediate precipitation. The results for different f values are shown in Fig. 3. It is obvious that the Rayleigh process leads to much higher fractionation than the slight temperature dependence of α in $\delta D_{evap.}$ as obtained by eqs. (3) and (4). This does not, however, take into consideration the effects of air mass mixing during transport. Therefore, it is difficult to determine the origin of air moisture only by its isotopic composition. The observed δD values between 20°S and 60°S correspond to an f value of 0.7 in Fig. 3. Air moisture samples at 65°S have an average δD value of about −170‰. According to the Rayleigh model (eq. 3) this value corresponds to a fraction f of 0.5 (Fig. 3). This means that only 50% of the original water vapour from the source region arrived this latitude. In nature however, isotopic compositions change not only as a result of condensation and precipitation, but also as a result of dynamic mixing during transport towards higher latitudes.

The distribution of deuterium in atmospheric water vapour in high latitudes (Fig. 3) might be explained by the meridional circulation of air masses as discussed by SCHWERDTFEGER (1984), where the inflow of air over polar ice caps occurs above the atmospheric boundary layer. The average isotopic composition δD at Mizuho Station (71°S) is about −166‰ at an average air temperature of −21.4°C. Similar δD values were measured at higher temperatures above the sea surface at 65°S. So it might be assumed that these isotopically depleted air masses reach the ice-covered continent at higher altitudes, or at least, that the air at Mizuho Station is less affected by condensation and precipitation than at Syowa Station. The δD average at Mizuho Station is higher than those from Syowa Station and Dome Fuji Station. The air moisture reaching Dome Fuji Station is isotopically lighter then at Mizuho Station, due to further water depletion by adiabatic cooling of the ascending air and subsequent precipitation, but the δD confidence interval indicates a strong scattering of the few (only 7) individual January observations (Fig. 3). According to HOLTON *et al.* (1995), this might also be explained by possible air mass inflow from upper troposphere or lower stratosphere due to subsidence in the circumpolar trough and tropopause folds, coupled with changing wind directions at an average January temperature of −35.3°C at Dome Fuji Station (SHIRAIWA *et al.*, 1996).

No prevailing wind direction could be observed in this region (Fig. 4), as was the

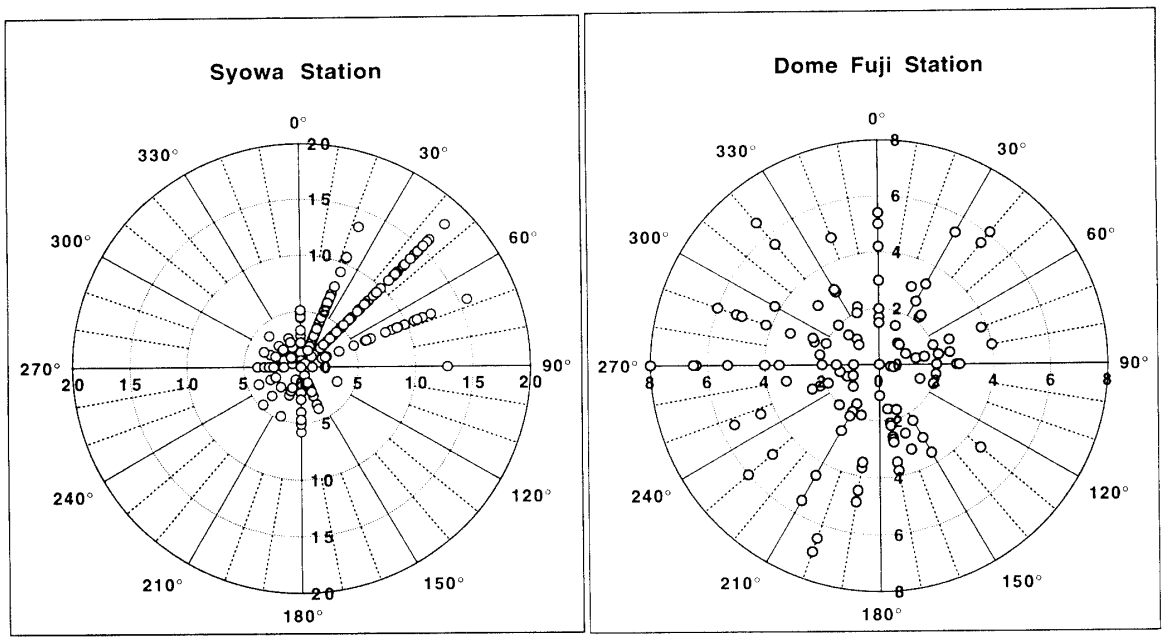

Fig. 4. Distribution of wind speed in m/s and wind direction in degrees at Syowa Station and at Dome Fuji Station during January 1995. Data are from SHIRAIWA *et al. (1996) and from JARE members of the Japan Meteorological Agency. Pay attention to the different wind speed scales !*

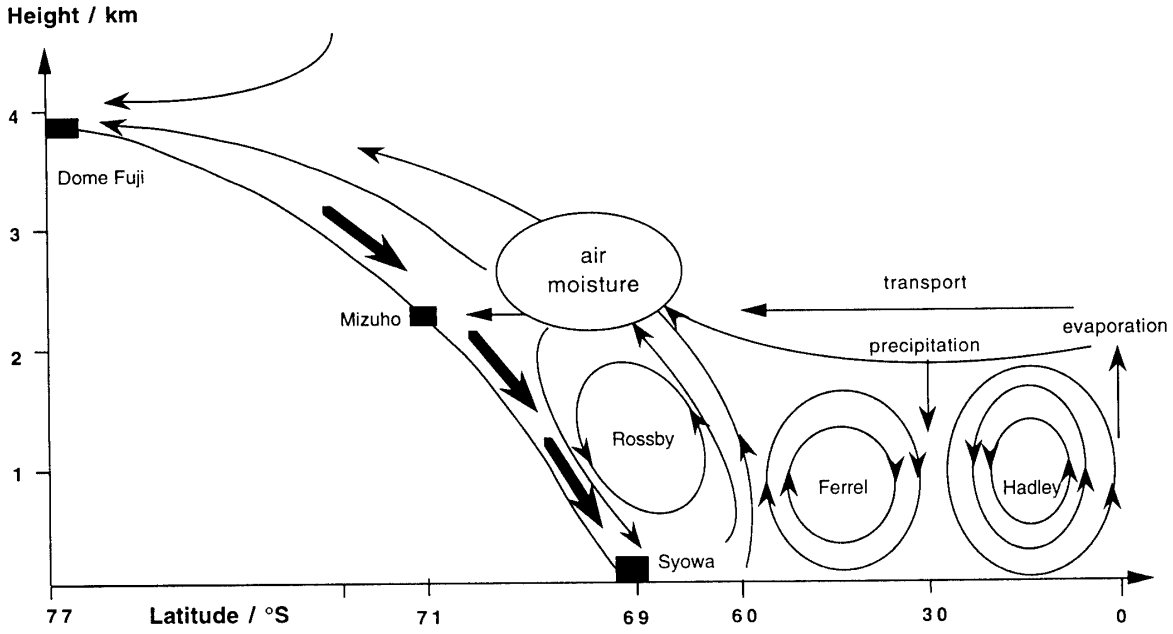

Fig. 5. Principal pattern of meridional circulation controlling the latitudinal isotopic distribution. Bold arrows symbolise strong katabatic winds.

case on the ice slope area (see Fig. 2 in ENOMOTO *et al.*, 1995) and at Syowa Station (Fig. 4). On the other hand, air is transported downward via Mizuho Station to Syowa Station by the prevailing cold and dry katabatic surface winds (ENOMOTO *et al.*, 1995;

SCHWERDTFEGER, 1984), marked with bold arrows in the simple circulation pattern in Fig. 5. Contrary to ENOMOTO *et al.* (1995), who claimed that no prevailing wind direction could be observed at Syowa Station throughout 1993, frequent strong winds were only recorded from north-easterly directions in January 1995 (Fig. 4). The δD values are very low in these air masses. During transport over the snow/ice surface, mixing with sublimated vapour seems to be an additional process contributing to the production of isotopically lighter air moisture. The observation of a minimum δD average at Syowa Station is not consistent with the so called altitude and latitude effects (HOEFS, 1987), where lower isotopic ratios appear at higher altitudes and at higher latitudes.

4. Conclusions

The deuterium distribution in atmospheric water vapour between 20° N and 60° S is strongly related to geographic position, due to the fact that latitude is closely correlated with temperature. Above 65° S δD values decrease more rapidly than temperatures, probably because of water depletion in the air masses by precipitation. A significant δD minimum was identified at 69° S (Syowa Station). This fact can been explained by the prevailing dry katabatic winds combined with mixing of sublimated vapour from the snow/ice surface. The average δD value at the continental Mizuho Station (71° S) is close to the observations from around 65° S, indicating that these samples may come from an air mass with a similar transport and water depletion history. This is consistent with the theory that the inflow of wet air masses to the Antarctic continent occurs above the atmospheric boundary layer. For Dome Fuji Station (77° S), different air flow processes are under discussion. Further cooling and precipitation may occur during upward and southward water vapour transport, and it is even possible that air mass inflow occurs as a result of subsidence of higher layers in the circumpolar trough. This probably contributes to the fact that the average δD at Dome Fuji Station has a large associated error, and is lower than at Mizuho Station, but not as low as at Syowa Station.

A preliminary conclusion can be drawn about air circulation patterns from general theories and the presented isotope study results, illustrated by the simple diagram in Fig. 5.

The source region of atmospheric water vapour is relatively less important for the final isotopic composition of Antarctic air moisture, because a slight temperature dependence of the fractionation factor α leads to small isotopic variations in contrast to the drastic isotope fractionations, caused by condensation, precipitation, exchange and mixing with other air masses during the long transport to Antarctica.

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